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STRUCTURE OF AROMATIC COMPOUNDS.

Part I, Acenaphthene.

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ABSTRACT.—From an analysis of the estimated intensities of X-ray reflections from a number of planes and by combining the available magnetic data, it is found that the two fold axis of the molecule lies parallel to the b-axis while the molecular plane is inclined at 26° to the c-face. The estimated intensities are satisfied by the assumption of a plane structure of the molecule The distance between an aliphatic carbon atom and its adjacent aromatic carbon atoms is 1.47A°, and that between the two aliphatic carbon atoms is 2.01A°. The angle between the aliphatic bond and the aliphatic-aromatic bond is 97° and angles between the latter bond and the adjacent bonds in the naphthalene nucleus are 114° and 126°.

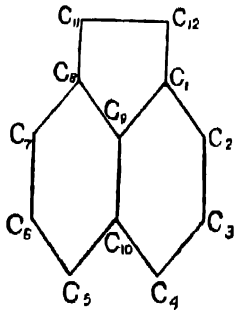
INTRODUCTION.

The high rigidity of the benzene ring in aromatic compounds is a very useful guide in the complete determination of the structures of such substances. These rings may be considered as separate units. Their size and shape as well as the arrangements of the carbon atoms inside them are known completely from the determinations of Lonsdale[1] and have been shown to remain unchanged in the condensed rings of naphthalene and anthracene[2] as well by one of the present authors. The benzene ring has been found to occur with its characteristic shape and size in all the aromatic crystals studied later on except in benzoquinone [3] in which the valency bonds inside the benzene ring are altered due to substitution of hydrogen by divalent oxygen. For determining the structures of the more complicated aromatic compounds, it is therefore possible to start with the known structure of the benzene nucleus and thereby the problem is simplified considerably.

Another very important guide to the structures of aromatic crystals is the magnetic property. From experiments on optical scattering and magnetic birefringence it was known that the benzene molecule has a very high optical

as well as magnetic anisotropy. One of the present authors found [2] that the optical and magnetic anisotropies in naphthalene and anthracene crystals measured by Bhagavantam[4] conform nicely with the structures determined from X-ray measurements. This magnetic method was found by Krishnan and his collaborators [5] to be extremely helpful in structure analysis of aromatic compounds. In a series of communications we propose to publish the results of a systematic study of the structures of aromatic organic compounds in which the data of our X-ray measurements are combined with the magnetic properties of the crystals as measured by Krishnan and others or by ourselves, and thereby to determine their atomic arrangements.

In this paper we shall present the results of structure analysis of acenaphthene. The carbon skeleton in the structural formula usually put forward by chemists is given below.



The two aliphatic carbon atoms (11) and (12) are connected by a single bond. In aliphatic compounds, however, we khow that the angle between two bonds is the tetrahedral angle of nearly 109° and the C-C distance is 1.54A° but as drawn here the angle between C_8C_{11} and $C_{11}C_{12}$ is apparently a right angle and the distance $C_{11}C_{12}$ appears to be much greater than 1.54A°. But if the aliphatic distance is to be preserved between these two carbon atoms the bonds C_8C_{11} and C_1C_{12} should be inclined from the symmetrical position. So it is interesting to see whether the atoms lie in the symmetrical positions, or in positions demanded by the aliphatic C-C distance or in positions different from both.

Space-group of Acenaphthene. Acenaphthene crystals belong to the orthorhombic bipyramidal class. The axial lengths as determined by us are

$$a_0 = 8.32A^\circ \; ; \; b_0 = 14.15A^\circ \; ; \text{ and } c_0 = 7.26A^\circ$$

and the number of molecules per unit cell is 4. These values are in agreement with those determined by W. H. Bragg. The space-group was found to be $D^5_{2h}p\gamma\mu\mu$ previously by Hertel and Kleu [6] from a few X-ray reflections. As we have an oscillation camera of high resolving power (diameter 17.18 cm.) it became possible for us to identify an extremely large number of reflecting planes and thereby to determine the space-group unequivocally. A list of the reflecting

planes thus identified are given in Table I. It will be seen from the table that the 'ausloschungen' observed by Hertel and Kleu are completely verified, *viz.*, that planes of the type o k l with l odd are absent. There are no other systematic 'ausloschungen.' This shows that the be plane is a glide plane of symmetry with glide component along the 'c' axis, the other two axial planes being ordinary planes of symmetry.

As there are four molecules in the unit cell the possible molecular symmetry according to Astbury and Yardley's [7] tabulated results on space-groups, is either a two fold axis normal to 010 or a plane of symmetry parallel to 010 or 001.

TABLE I.

List of reflecting planes for acenaphthene
(Oscillation about 'b' and 'c' axes).

Axial planes.

Plane.	Intensity.	Plane.	Intensity.
0 0 2	v.s.	0 9 0	v.w.
0 0 4	s.	0 10 0	w.
0 0 6	s.	0 11 0	m.
0 3 0	m.s.	2 0 0	s.
0 4 0	v.s.	3 0 0	m.s.
0 7 0	m.w.	4 0 0	m.w.
0 8 0	v.s.	5 0 0	m.s.
		6 0 0	v.v.w.

Prism Planes.

Plane.	Intensity.	Plane.	Intensity.
0 1 2	v. s.	0 3 2	m.
0 1 4	v. s.	0 3 4	m.
0 1 6	m.	0 3 6	v. w.
0 2 2	m.	0 4 2	w.
0 2 4	m.	0 4 6	w.
0 2 6	w.	0 5 2	s.

Prism Planes (contd.).

Plane.	Intensity.	Plane	Intensity.
0 5 6	m.	4 0 4	m.
0 6 2	v.s.	4 0 5	m.
0 7 2	s.	4 0 6	m.
0 8 2	m. w.	4 0 7	m.
0 9 2	m. w.	5 0 1	s.
0 10 2	w.	5 0 2	m.
0 11 2	m. w.	5 0 3	m.
0 12 2	m.	5 0 4	m.
1 0 1	s.	5 0 5	m.w.
1 0 2	v. s.	5 0 7	m.w.
1 0 3	s.	6 0 1	w.
1 0 4	s.	6 0 2	m.
1 0 5	s.	6 0 3	m.
1 0 6	m. w.	6 0 4	m.w.
1 0 7	m. w.	6 0 5	m.
2 0 1	v.s.	7 0 1	v.w.
2 0 2	s.	7 0 2	m.
2 0 3	s.	7 0 3	m.
2 0 4	m.	7 0 4	m.
2 0 5	m.	8 0 1	m.w.
2 0 6	m.w.	8 0 2	w.
3 0 1	s.	8 0 3	m.w.
3 0 2	s.	8 0 4	m.w.
3 0 3	m.	9 0 2	w.
3 0 4	w.	9 0 3	w.
3 0 5	v.w	1 2 0	v.s.
3 0 6	w.	1 3 0	s.
4 0 1	s.	1 4 0	m.
4 0 2	s.	1 5 0	m.w.
4 0 3	m.	1 6 0	m.w.

Prism Planes (contd.).

Plane.	Intensity.	Plane.	Intensity.
1 7 0	w.	3 13 0	v. v. w.
1 8 0	m.	4 1 0	m.
1 9 0	w.	4 1 0	m.
1 10 0	m.w.	4 3 0	v. w.
1 11 0	m.w.	4 4 0	v. w.
1 13 0	w.	4 5 0	w.
1 14 0	v. v. w.	4 6 0	m. w.
2 1 0	s.	4 7	m.
2 3 0	m.	4 8 0	w.
2 4 0	m. w.	4 9 0	v. v. w.
2 5 0	m.	4 10 0	v. v. w.
2 6 0	s.	4 12 0	v. w.
7 0	m.w.	4 13 0	v. w.
2 8 0	v. w.	4 14 0	v. w.
2 10 0	v. w.	5 1 0	m.
2 11 0	v. w.	5 2 0	m. w.
2 12 0	w.	5 3 0	w.
2 13 0	w.	5 4 0	m.
3 1 0	m.	5 5 0	m. w.
3 2 0	s.	5 6 0	s.
3 3 0	v. v. w.	5 7 0	w.
3 4 0	m. w.	5 10 0	w.
3 5 0	s.	5 11 0	w
3 6 0	m.	5 13 0	w.
3 7 0	w.	5 14 0	w.
3 8 0	v. w.	6 1 0	m. w.
3 9 0	v. v. w.	6 2 0	m. w.
3 10 3	m. w.	6 3 0	w.
3 11 0	w.	6 4 0	v. v. w.
3 12 0	m. w.	6 5 0	m. w.

Prism Planes (contd.).

Plane.	Intensity.	Plane.	Intensity.
6 6 0	v. v. w.	7 11 0	w.
6 7 0	v. v. w.	8 2 0	m.
6 9 0	v. w.	8 3 0	v. w.
7 1 0	m.	8 6 0	v. w.
7 3 0	m. w.	9 4 0	v. w.
7 5 0	v. v. w.	9 6 0	m
7 6 0	m. w.	10 4 0	w.
7 10 0	w.		

General Planes

Plane.	Intensity.	Plane.	Intensity.
1 1 1	v. s.	1 5 2	s.
1 1 2	s.	1 5 3	v. s.
1 1 3	m.	1 5 4	m.
1 1 4	m. w.	1 5 5	v. w.
1 1 5	w.	1 6 1	m. w.
1 1 6	v. v. w.	1 6 2	m. w.
1 2 1	m.	1 6 3	w.
1 2 2	m.	1 6 4	w.
1 2 3	v. w.	1 6 5	s.
1 2 6	v. v. w.	1 6 6	s.
1 3 1	v. s.	1 6 7	w.
1 3 2	m.	1 6 8	w.
1 3 3	m. w.	1 7 1	s.
1 4 2	m. w.	1 7 2	m. w.
1 4 3	s.	1 7 3	m.
1 4 4	v. v. w.	1 8 1	m. w.
1 5 1	v. s.	1 8 2	m. w.

General Planes (contd.).

Plane.	Intensity.	Plane.	Intensity.
1 8 3	v. v. w.	2 4 2	s.
1 9 1	w.	2 4 3	v. w.
1 9 2	m. w.	2 4 4	m. w.
1 10 1	m. w.	2 4 5	w.
1 10 2	v. w.	2 4 6	v. v. w.
1 10 3	m. w.	2 4 7	v. w.
1 11 1	s.	2 5 1	m.
1 11 2	w.	2 5 2	m. w.
1 11 3	w.	2 5 3	m.
1 12 2	w.	2 5 4	s.
1 12 3	m.	2 5 5	m.
1 13 1	m. w.	2 5 6	m.
2 1 1	v. s.	2 5 7	v. w.
2 1 2	s.	2 5 8	m. w.
2 1 3	m.	2 6 1	v. s.
2 1 4	m. w.	2 6 2	v. s.
2 1 5	m.	2 6 3	v. s.
2 1 8	v. w.	2 6 4	s.
2 2 1	v. s.	2 6 5	w.
2 2 2	m. w.	2 6 7	v. w.
2 2 3	s.	2 7 1	m.
2 2 4	v. w.	2 7 2	w.
2 2 5	w.	2 7 3	m. w.
2 3 1	m.	2 8 1	m.
2 3 2	w.	2 8 2	v. w.
2 3 3	m. w.	2 8 3	m. w.
2 3 4	v. w.	2 9 1	w.
2 3 5	w.	2 10 1	v. w.
2 3 7	v. v. w.	2 10 5	v. w.
2 4 1	m.	2 11 1	v. w.

General Planes (contd.).

Plane.	Intensity.	Plane.	Intensity.
2 11 2	v. w.	3 4 5	w.
2 11 3	v. w.	3 4 6	v. w.
2 12 1	w.	3 5 1	v. s.
2 12 2	w.	3 5 2	s.
2 12 3	w.	3 5 3	m.
2 13 1	v. w.	3 5 4	m.
2 13 2	v. w.	3 5 5	w.
2 14 2	v. w.	3 6 1	w.
2 14 3	v. w.	3 6 2	
3 1 1	m.	3 6 3	m. w.
3 1 2	s.	3 6 4	m.
3 1 3	s.	3 6 5	m. w.
3 1 4	v. w.	3 6 6	w.
3 1 5	v. w.	3 6 7	v. w.
3 1 6	v. w.	3 7 1	m. w.
3 2 1	m. w.	3 7 2	v. w.
3 2 2	v. w.	3 7 3	m.
3 2 3	v. v. w.	3 8 3	w.
3 2 4	v. w.	3 9 1	m. w.
3 3 1	m.	3 9 2	m. w.
3 3 2	m.	3 10 1	w.
3 3 3	v. w.	3 10 2	w.
3 3 4	w.	3 10 3	w.
3 3 5	w.	3 11 1	w.
3 3 7	v. w.	3 11 2	m. w.
3 3 8	m.	3 11 3	v. w.
3 4 1	m.	3 12 1	m. w.
3 4 2	m. w.	3 12 2	m. w.
3 4 3	m. w.	3 12 3	w.
3 4 4	w.	3 13 1	v. w.

General Planes (contd.).

Planes.	Intensity.	Planes.	Intensity.
3 13 2	v. w.	4 7 3	w.
3 13 3	w.	4 8 1	w.
4 1 1	m.	4 8 2	v. w.
4 1 2	s.	4 9 1	v. v. w.
4 1 3	s.	4 9 2	v. w.
4 1 4	m.	4 9 3	v. w.
4 1 5	w.	4 10 2	m. w.
4 2 1	m. w.	4 11 1	w.
4 2 2	m.	4 11 2	m. w.
4 2 4	w.	4 11 3	w.
4 2 5	v. w.	4 12 2	w.
4 2 6	v. w.	4 12 3	v. w.
4 3 1	m, w.	4 13 1	v. v. w.
4 3 2	w.	4 13 2	w.
4 3 3	v. w.	4 13 3	w.
4 3 4	w.	5 1 1	m.
4 4 1	w.	5 1 2	m. w.
4 4 2	v. w.	5 1 3	m. w
4 4 4	v. w.	5 1 4	v. w.
4 5 1	m.	5 1 5	v. w.
4 5 2	v. w.	5 1 6	v. w.
4 5 3	v. w.	5 2 2	v. v. w.
4 5 4	m. w.	5 2 3	m. w.
4 5 7	v. w.	5 2 4	v. v. w.
4 6 1	m.	5 3 1	m. w.
4 6 2	m. w.	5 3 3	v. w.
4 6 3	w.	5 3 4	w.
4 6 4	m. w.	5 3 5	v. w.
4 6 6	m, w.	5 4 1	v. w.
4 7 1	w.	5 4 5	v. w.

General Planes (contd.).

Planes.	Intensity.	Planes.	Intensity.
5 4 6	v. w.	5 13 1	v. w.
5 5 1	v. w.	6 1 1	m.
5 5 2	s.	6 1 2	m. w.
5 5 3	m. w.	6 1 3	m. w.
5 5 4	m. w.	6 1 4	v. w.
5 5 5	v. v. w.	6 1 5	v. w.
5 5 6	v. w.	6 2 1	w.
5 5 7	w.	6 2 2	v. w.
5 6 1	m.	6 2 3	m.
5 6 2	s.	6 2 5	v. v. w.
5 6 3	m.	6 3 1	m. w.
5 6 4	w.	6 3 2	m. w.
5 6 5	w.	6 3 3	v. v. w.
5 6 6	w.	6 3 4	v. w.
5 7 1	m. w.	6 3 5	v. v. w.
5 7 2	m. w.	6 4 1	v. w.
5 7 3	w.	6 4 2	v. w.
5 8 1	m. w.	6 4 3	m. w.
5 8 2	w.	6 5 1	s.
5 8 3	m. w.	6 5 2	m. w.
5 9 1	m. w.	6 5 3	m. w.
5 9 2	w.	6 5 4	m. w.
5 10 1	v. w.	6 5 5	m.
5 10 2	m. w.	6 6 1	m.
5 10 3	m. w.	6 6 2	m. w.
5 11 1	w.	6 6 3	m.
5 11 3	w.	6 6 4	w.
5 12 1	w.	6 6 5	m. w.
5 12 2	w.	6 6 6	m. w.
5 12 3	w.	6 7 1	m. w.

General Planes (contd.)

Planes.	Intensity.	Planes.	Intensity.
6 7 2	w.	7 5 4	w.
6 7 3	w.	7 6 1	m. w.
6 8 1	v. v. w.	7 6 2	w.
6 8 3	w,	7 6 3	m. w.
6 9 2	v. v. w.	7 6 4	w.
6 9 3	v. v. w.	7 7 1	v. w.
6 11 1	v. v. w.	7 7 2	w.
6 11 2	w.	7 8 2	w.
6 11 3	w.	7 9 2	w.
6 12 1	w.	7 10 3	w.
6 12 2	w.	7 11 1	v. v. w.
7 1 2	v. w.	8 1 1	w.
7 1 3	w.	8 1 2	w.
7 1 4	v. w.	8 1 4	m. w.
7 1 6	v. w.	8 2 2	v. v. w.
7 2 1	v. w.	8 3 1	v. w.
7 2 2	v. w.	8 4 1	v. v. w.
7 2 3	w.	8 4 2	v. w.
7 2 4	v. w.	8 5 1	v. v. w.
7 3 1	w.	8 5 2	m. w.
7 3 2	v. w.	8 5 3	w.
7 3 3	v. v. w.	8 6 1	v. w.
7 3 4	v. w.	8 6 2	m. w.
7 4 1	v. w.	8 6 3	v. w.
7 4 2	v. w.	8 7 1	v. w.
7 4 3	v. v. w.	8 7 2	s.
7 4 4	v. v. w.	8 8 2	s.
7 5 1	v. w.	9 1 2	v. w.
7 5 2	m. w.	9 4 1	v. w.
7 5 3	m. w.	9 4 2	w.

General Planes (contd.).

Plane.	Intensity.	Plane.	Intensity.
9 5 1	v. w.	10 1 2	v. w.
9 5 2	m. w.	10 2 2	v. w.
9 6 1	m. w.	10 4 1	v. w.
9 6 2	w.		

Atomic Arrangement in Acenaphthene.

As we have seen before from its space-group, acenaphthene molecule must possess either a plane of symmetry parallel to the ' b ' or the ' c ' face or an axis of symmetry normal to the ' b '-face. Thus we have to try three classes of arrangements and let us call them respectively as class ' A,' class ' B ' and class ' C.' From the stereochemical formula of acenaphthene it is seen that the two-fold axis if there be one should pass through C_9C_{10}. On the other hand if the molecule possesses a plane of symmetry, the plane of symmetry must be either the plane containing the naphthalene nucleus or the plane normal to that through C_9C_{10}.

Class A.—*The molecule possessing a plane of symmetry parallel to the ' b '-face.* Two cases may arise according to whether the plane of the naphthalene nucleus or its normal is the plane of symmetry. In the first alternative, the plane of the molecule is the ' b '-face. In that case the diamagnetic susceptibility along the ' b ' axis should be the maximum, but on the contrary it is the minimum as found by Krishnan, Guha and Banerjee.[5] So we may discard this type of orientation as untenable. According to the other case in this class the length of the molecule should be parallel to the ' b ' axis as it is normal to the plane of symmetry. The gram molecular diamagnetic susceptibility in this direction should be equal to that of naphthalene along the length of the molecule but for the modification due to the two CH_2 groups at the side. It is highly improbable that only the two CH_2 groups at the side can modify the gram molecular susceptibility in this direction from $-39{\cdot}4$ to $-72{\cdot}1$. There is another difficulty which makes this orientation impossible. In acenaphthene crystal all the three axial planes are planes of symmetry. In the orientation that we are considering, since the ' b ' plane is a molecular plane of symmetry the other two planes of symmetry are brought about by repetitions of molecules. So we have to accommodate cross-sections of four molecules within the ' b'-face of the unit cell which is too small for that. Thus the possibility of a plane of symmetry parallel to the ' b '-face is precluded.

Class 'B'.—*The molecule possessing a plane of symmetry parallel to the 'c'-face.* Here also as before two cases arise. The first case is that the molecular plane is parallel to the 'c'-face. The gram molecular susceptibility in this case should be maximum along 'c' and its value should not be less than that for the direction normal to the naphthalene molecule since the average value (111·8) for the molecule is much greater than that for naphthalene (89·8). But though it has been found to be the maximum along the 'c' axis its value is $-145{\cdot}6 \times 10^{-6}$ while that for normal to the naphthalene molecule is $-187{\cdot}2 \times 10^{-6}$. The other case is precluded by the fact that in this case the length of the molecule should be along the 'c' axis and consequently the molecular susceptibility should be the minimum while actually it is the maximum.

Class 'C'.—Thus we are left with the only possibility that the 'b' axis is a two fold axis of symmetry of the molecules so that it coincides with C_9C_{10}. In this direction it is probable that the susceptibility of the molecule is altered to some extent from the value for the naphthalene nucleus due to the presence of the two CH_2 groups. As regards the susceptibilities in the other directions agreements may be obtained by rotating the molecule to the requisite amount about the two fold axis.

The mean gram molecular susceptibility of acenaphthene is $-111{\cdot}8 \times 10^{-6}$ Let the gram molecular susceptibilities along the length, breadth and thickness of the molecule be respectively K_1, K_2 and K_3 then

$$K_1 + K_2 + K_3 = -335{\cdot}4 \times 10^{-6}.$$

K_2 is the value along the 'b' axis, so that it is $-72{\cdot}1 \times 10^{-6}$. Hence $K_1 + K_3 = -263{\cdot}3 \times 10^{-6}$. The value of the corresponding quantity for naphthalene is $-226{\cdot}6 \times 10^{-6}$.

So if we consider that the CH_2 groups influence K_1 and K_3 equally, we have

$$K_1 = -57{\cdot}7 \times 10^{-6} \quad \text{and}$$

$$K_3 = -205{\cdot}5 \times 10^{-6}.$$

Suppose the molecular plane makes an angle α with the 'c'-face then we may write out the following equation correlating the susceptibilities.

$$117{\cdot}6 = 57{\cdot}7 \cos^2\alpha + 205{\cdot}6 \sin^2\alpha$$

whence

$$\alpha = 39^\circ\ 30'$$

We have taken from the known structures of the other aromatic compounds the distance between consecutive carbon atoms in the benzene rings to be 1·42 A°, while those between $C_1 - C_{12}$ and $C_{11} - C_{12}$ have been determined by trial. At first the intensities of the axial planes were calculated for the molecular plane at $39\frac{1}{2}°$ with the ' c '-face. By altering the C_1C_{12} and $C_{11}C_{12}$ distances and the position of the molecule along the ' y '-axis within reasonable limits, it was not possible to get agreement with the estimated intensities. The angle α and these lengths as well as the position of the molecule along the ' b '-axis were then varied until the best agreement could be obtained. For this the angle α is 26° and the distance C_1C_{12} and $C_{11}C_{12}$ are respectively 1.47 A° and 2.01 A°. The atomic parameters for half of a molecule are given in Table II.

TABLE II.

Atomic Parameters in Acenaphthene Crystal.

Atoms.	x.	y.	z.
C_1	0·131	·280	·072
C_2	·261	·239	·144
C_3	.261	·139	·144
C_4	·131	·089	·072
C_9	0	·239	0
C_{10}	0	·139	0
C_{12}	·111	·400	·061

The nature of agreement for the axial planes can be seen from the Table III, in which the first column gives the planes, the second column gives calculated structure factors and the third column gives the estimated intensities. For the calculations, the atomic structure factors for the aromatic carbon atoms were taken from the table given by Robertson (*Proc. Roy. Soc.*, Vol. 150A, p. 110, 1935) and for the aliphatic carbon atoms, the values given by Ponte for diamond were adopted (*Phil. Mag.*, Vol. 3, p. 195, 1927).

From the above discussions we find that the distance $C_8C_{11} = C_1C_{12} = 1{\cdot}47A°$, and $C_{11}C_{12} = 2.01A°$. The angle $C_8C_{11}C_{12}$ = the angle $C_1C_{12}C_{11} = 97°$ and the angle $C_9C_8C_{11}$ = the angle $C_9C_1C_{12} = 114°$. Thus we see that the bonds C_8C_{11}

TABLE III.

Calculated and observed Intensities of Reflection.

Indices.	Calculated Structure Factor.	Estimated Intensities.
200	− 26·2	s
300	− 11·7	ms
400	− 7·2	mw
500	− 32·3	ms
600	− 3·4	vvw
030	− 19·4	ms
040	− 32·2	vs
050	− 11·6	Not observed.
060	5·9	
070	− 11·1	mw
080	41·8	vs
090	− 15·5	vw
0(10)0	8·3	w
0(11)0	− 19·0	m
002	72·3	vs
004	17·2	s
006	− 16·1	s

and C_1C_{12} have properties intermediate between aliphatic and aromatic carbon atoms, both as regards angle and size, while the purely aliphatic bond $C_{11}C_{12}$ has been lengthened out.

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